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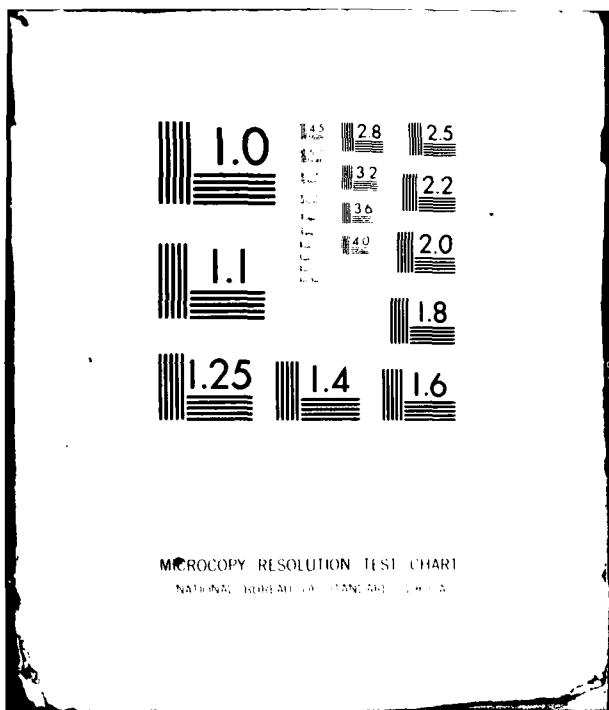
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(6) MOLECULAR LEVEL MODELING OF THE MECHANISM OF
SHOCK INITIATION OF SOLID EXPLOSIVES (1)

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INTRODUCTION: The ability to control the initiation of explosives is essential to both military and civilian use of the materials. Control in the past has been exercised by empirical reformulation of composition in order to adjust sensitivity. There is at present little fundamental understanding on the molecular level of the phenomena of shock initiation, a situation which can only have a detrimental effect on the possibility of attainment of finely controllable insensitive high explosives.

Since explosion is a chemical reaction and because the quantum theory has made abundantly clear that the reactive properties of a material are determined by the vibrational and electronic structure of the constituent molecules or atoms of the substance, understanding of initiation to detonation will only emerge by approaching the problem on a fundamental molecular level. An important objective of such an approach would be the establishment of a relationship between the useful properties of explosives such as sensitivity and yield and their molecular properties.

This work represents an attempt to consider on a fundamental level how shock or impact can cause chemical reactions in certain solids in order to gain insight into the factors which determine sensitivity to explosion.

GENERAL MODEL: A detonation is a high rate, self-sustaining exothermic chemical reaction. One plausible model for the process envisions the shock pulse breaking intramolecular bonds of the constituent molecules of the solid explosive producing reactive

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fragments such as radicals. Since this step is endothermic, the necessary exothermic reactions are viewed to be among the secondary products created by the shock pulse. Detonation is achieved when a sufficient number of these products are present in order that enough energy is liberated to cause further reaction enabling the process to become self-sustaining. The first evidence that the initial step is endothermic and requires a build up of secondary products has previously been described by this author and collaborators (1,2). Solid explosives were subjected to shock pulses less than necessary to cause initiation to detonation and examined by electron paramagnetic resonance and x-ray photoelectron spectroscopy. The results showed definitively that a preliminary build up of radicals occurred and specifically identified the bonds broken. In the case of triamino trinitrobenzene (TATB) and 1,3,5 trinitro-1,3,5 triazacyclohexane (RDX), NO_2 was severed from the ring of the molecule. In ammonium nitrate (AN) the NH_4^+ ion was observed to decompose by shock.

This paper is concerned with understanding on a molecular level how a shock pulse causes high rate chemical reactions in explosive solids. There are two general ways a shock pulse can break a bond of a molecule in a solid. One process can be characterized as thermal, where the temperature generated by shock compression of lattice is the parameter which determines the reaction. The lattice heat is transferred to the molecules of the material causing bonds of the molecules to break. More recently the idea has been considered that the chemistry is a result of mechanical processes (3). Because of abrupt changes in potential molecules are removed from their lattice site colliding with nearest neighbors causing molecules to dissociate. To some extent, as will be seen below, when specific mechanisms are considered this distinction between thermal and mechanical is artificial. Indeed examination of available macroscopic experimental data for secondary explosives suggests the chemistry induced by the shock pulse is a result of heat generated by shock compression. Inhomogeneous explosives, where the temperature rise is greater for a given shock pressure, are much more sensitive to shock than homogeneous single crystals. Also, as shown in Figure 1, there is a correlation between impact sensitivity and thermal sensitivity. Thermal initiation of explosion is generally understood in terms of the heat conduction equation

$$\rho C \frac{\partial T}{\partial t} = \tau \nabla^2 T = Q N K \quad (1)$$

T being temperature, C the specific heat, ρ the density, τ thermal conductivity, Q heat of reaction, and K rate of reaction. The solution of the equation predicts a time delay, τ , at given temperature

OWENS

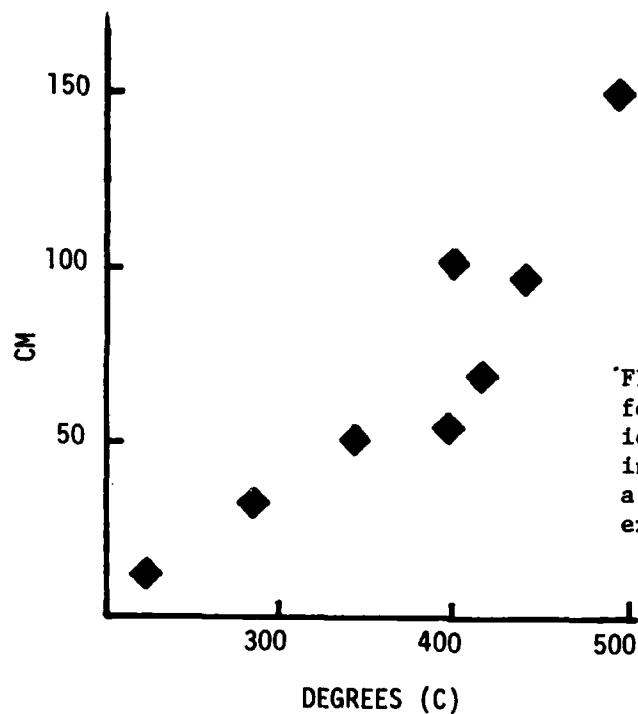


FIGURE 1. Impact height for initiation to detonation versus five second initiation temperature for a number of secondary explosives.

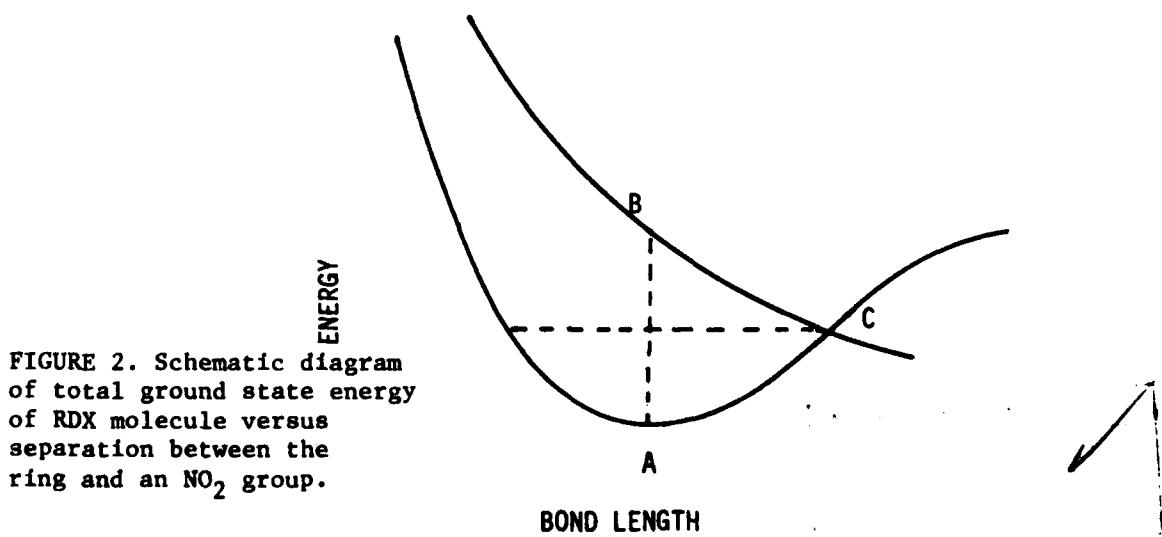


FIGURE 2. Schematic diagram of total ground state energy of RDX molecule versus separation between the ring and an NO_2 group.

from the time the temperature is turned on to explosion. This delay time depends on temperature.

$$\tau = Ce^{-E/kT} \quad (2)$$

Numerous measurements of delay times have been recorded and the data is described by equation (2). More recently delay times have been measured after application of shock pulses needed to detonate single crystals of RDX and PETN (4). This data is shown in Table I. These delay times after application of shock are predicted from thermal explosion theory by extrapolation of thermal delay time data described by equation (2) to the temperatures calculated at the critical shock pressure. This suggests that the reactions of shock initiation are indeed the same as thermal initiation.

Table I. Results of Measurement of Time to Detonation
After Application of Shock Pulse of Peak
Pressure Equal to Critical Pressure in
Single Crystals of Explosive Solids*

| <u>Explosive</u> | <u>Critical Pressure (Kbs)</u> | <u>Time to Explosion (s⁻¹)</u> |
|------------------|--|---|
| PETN | 112 | 3×10^{-7} |
| RDX | 170 | 1×10^{-6} |
| Tetryl | 150 | 1×10^{-6} |
| TNT | 200 | 7×10^{-7} |

*Ref 4

The activation energies for thermal initiation deduced from equation (2) do not in general agree with activation energies measured from slow thermal decomposition experiments indicating that in the temperature regime of initiation different chemical reactions may be dominant than at lower temperatures. This result is also true of shock induced chemical reactions. Experiments in this laboratory which have identified the bonds broken by shock have shown that different bonds were broken in slow thermal decomposition (1,2). For example, in TNT heating to temperatures below initiation temperatures removes one oxygen of the nitro group whereas a shock pulse

OWENS

appeared to alter the oxygen-hydrogen bonding on the molecule. The evidence then is that in secondary explosives the shock induced reactions are a result of heating of the lattice. However, the reactions are not necessarily the same as reactions measured at temperatures below initiation temperatures. Thus, it appears that shock induced chemical reaction is a thermal process and that the temperature generated by the shock pulse is the most important parameter in determining the reaction rate.

The work here considers possible ways a chemical reaction can occur in a crystal lattice with particular emphasis on shock induced chemical reactions. The work considers specific plausible molecular mechanisms for shock induced reactions in solids and calculates reaction rates in order to assess the feasibility of the mechanisms in initiation of detonation.

In order for an intramolecular bond to break, heat or mechanical energy of the shock pulse which is absorbed by the lattice must be transferred to the molecules. This "energization" of the molecule can occur by either an excitation of an electronic or vibrational state of the molecule. The energy states of the molecule in the short time of the shock compression are not necessarily in equilibrium with the lattice. There must be a coupling between the states of the molecule and the lattice states. The strength of this coupling could determine the sensitivity to initiation. The molecules of the lattice may also be energized by collisions with other molecules. However, because they are not as free to move as in a gas or liquid, the way these collisions occur in a solid will be quite different than in the liquid or gas phase.

In developing theoretical models for mechanisms of reaction in a shock compressed solid it is necessary to characterize the heating of the lattice due to the shock pulse. Only initiation of reaction in single crystals will be considered where the temperature rise is due to bulk heating. It will be assumed that the lattice states achieve equilibrium within a time short compared to the duration of the shock, typically 10^{-6} S. Without this assumption the concept of temperature would not be appropriate. On the other hand, it will not be assumed that the internal states of the molecule are in equilibrium with the lattice. In fact, the results of one model will justify this assumption. Therefore, in developing models for the reactive processes associated with internal states of the molecules nonequilibrium approaches will be necessary. In mechanisms involving external lattice states equilibrium theories may be appropriate.

Since temperature is a critical parameter in determining rate of reaction, modeling of reaction rates in shocked solids requires a calculation of temperature as a function of shock pressure. The calculation of shock temperature requires a knowledge of the equation of state and treats the material as a fluid characterized by only three thermodynamic variables. Examination of various approaches to calculating bulk shock temperatures indicates considerable lack of agreement in predictions suggesting that such calculations should be only regarded as estimates (5,6). Here a relatively simple method has been used where the temperature rise is given by, (7)

$$\Delta T = \frac{\Gamma T_0}{\rho C_p} \Delta P \quad (3)$$

where Γ is volume coefficient of expansion, T_0 initial temperature, ρ density, and C_p specific heat. For example, in RDX equation (3) yields a temperature of 585°K at 170 Kbs compared to 770°K calculated by Voskoboinikov (4). This is typical of the kind of agreement of various calculations.

THERMAL PROCESSES:

1. Vibrational Relaxational Processes: A shock pulse heats the solid by exciting vibrational states of the lattice. Three excited phonons are most likely to be transferred from the heated lattice to excite an internal vibration of one or more modes of the molecules which constitute the lattice. It has been shown for a Debye lattice that the rate of transfer, W_{10} , at a given temperature T to a bond of vibrational frequency ν is given by, (8)

$$W_{10} = \frac{9K N}{16\hbar m(4\pi M)} \frac{A T}{\nu^7} \quad (4)$$

where M is the mass of the molecule, m mass of atom on the molecule, ν_D is the Debye frequency of the lattice, A is a coupling constant between the lattice mode and an internal molecular vibration, K is Boltzman's constant, and \hbar is Planck's constant.

Unimolecular breaking of the bond may occur if there is a sufficient probability for multiple excitations to a vibrational state which has energy equal to or greater than the dissociation energy of the bond. With a knowledge of W_{10} , and assuming a harmonic oscillator potential it is possible to calculate the rate of excitation to the dissociative state using a random walk stochastic theory developed by

Montroll and Shuler (9). Application of the theory to the N-N bond breaking of RDX at temperatures appropriate to 170 Kbs of shock pressure indicates there is no possibility for this kind of a process. This calculation ignored coupling between internal modes of the molecule. This coupling which has a shorter relaxation time (10^{-9} S) than the molecule-lattice relaxation time (10^{-6} S) would compete with a multiple excitation process further reducing the probability of bond breaking.

The calculated W_{10} at 170° Kbs and 770° K is 1.3×10^{-4} S⁻¹ which means the relaxation time ($1/W_{10}$) for the transfer of phonons to the N-N vibration of RDX is longer than 10^{-6} S the typical pulse width used to initiate reaction. These results suggest that the internal vibrational states of the molecule do not have time to come to equilibrium within the duration of the shock pulse.

2. Electronic Excitation: Since it has been observed that UV light of wavelength which causes an electronic excitation can cause NO_2 to be severed from the ring of TATB and RDX, and since the same bond is broken by a shock pulse it is suggested that the first step in shock induced bond breaking is an electronic excitation. Recently French workers have indicated a correlation between the charge distribution calculated from molecular orbital theory of the excited state and impact sensitivity (10). The excited electronic structure of the RDX molecule has been elucidated by the work of Marinkas (11) and this work has shown that although there are a number of excited states, only the allowed excitations at 340 nm and shorter wavelength cause photolysis resulting in rupture on the R- NO_2 bond. Presumably this excited state is a dissociative state with respect to the N-N separation as schematically depicted in Figure 2. Direct thermal excitation of this level (line AB) is not probable at 770° K. However, thermal excitation to the unbound state could occur if the potential energy curve of this state as a function of N-N separation intersected the ground state potential energy (point C). Then thermal excitation of the ground state to point C would allow the possibility that the system cross over to the dissociative state leading to bond breaking. Since it is likely that this is the process for slow thermal decomposition point C the intersection point of the potential curves can be fixed at 2.0 eV for RDX which is the activation energy measured from slow decomposition studies (12). Using the formalism of equation (4) the rate at which the N-N vibration can be excited to a vibrational level 2.0 eV above the ground state which would be the 12th vibrational level can be calculated. The result is a rate again far too low compared to the rate needed to cause detonation, (10^6 S⁻¹). This mechanism, however, could be the

rate controlling step in slow thermal decomposition where lower reaction rates exist, but is excluded as a mechanism for shock initiation. This conclusion is consistent with the previous analysis which showed that extrapolation of slow thermal decomposition rates to critical shock temperatures gives rates which are too low to account for the process.

The elimination of thermal excitation to the dissociative state in RDX as the first step of shock induced bond breaking assumes that the separation between the ground and excited electronic state is not greatly affected by pressure. This assumption is justified by data on the effects of high pressure on the internal vibrational frequencies of the RDX molecule which will be presented below.

The elimination of the role of any internal vibrational or electronic states in breaking the N-N bond is really a result of the fact that W_{10} is too low meaning there is not time for energy to be transferred to internal states of the molecule during the shock compression.

3. Vacancy Production: Another possible way the chemical reactions of explosion may start in the solid is that the thermal energy generated by the shock pulse may cause molecules to be removed from their lattice sites to interstitial positions. In these interstitial positions they may be sufficiently close to neighboring molecules to allow chemical contact and thus reaction. The rate at which a molecule can be removed from its lattice site as a function of temperature can be approximated by

$$R = v_0 e^{-E/kT} \quad (5)$$

Where E is the energy needed to remove the molecule from its site to the interstitial position, and v_0 is the vibrational frequency of the molecule at its lattice site. In the case of ionically bonded explosives (such as ammonium nitrate) where electrical conductivity at elevated temperatures is determined by cation diffusion, E can be estimated from studies of the temperature dependence of the electrical conductivity to be 1.17 eV (13). The translational vibrational frequencies of the NH_4^+ ion in AN have been measured by Raman spectroscopy (14). Thus equation (5) and a calculation of the lattice temperature as a function of peak shock pressure enables the rate of removal of cations from lattice sites to be calculated. At a shock pressure of 100 Kbs a reaction rate of $8 \times 10^5 \text{ s}^{-1}$ is obtained which is in the order of magnitude of the rates needed to cause the initiation to detonation transition.

OWENS

This analysis has not taken into account the effect of pressure on v_0 or E. Raman studies of effect of pressure on lattice vibrations suggest v_0 would increase by about 1/2 % per kilobar (15). Electrical conductivity decreases by an order of magnitude for a two order of magnitude increase in pressure (16). These numbers indicate that at 100 Kbs E would increase roughly by 30% which would reduce the reaction rate by about an order of magnitude from the estimate that does not take into account lattice compression. Thus vacancy production remains a possibility for the initial step which produces reaction. Indeed vacancies have been observed to be produced in solids by shock (17). Significant suppression of reaction rates in explosive solids with increasing hydrostatic pressure has not been observed (18).

4. Large Amplitude Lattice Vibrations: An examination of measured lattice mode frequencies in a number of chemically similar primary explosives indicates a correlation between certain mode frequencies and sensitivity to thermal initiation, suggesting a possible role of lattice vibrations in the initiating mechanism. In gas or liquids one of the factors that determines bimolecular reaction rates is the probability that the two reacting molecules can come into chemical contact. In a solid where the motion of the ions is constrained by the normal modes of vibration of the lattice, there will be certain modes of vibration in which two adjacent molecules of the lattice approach each other. If the amplitude of vibration is sufficiently large at elevated temperatures the two ions may get close enough to chemically react. The rate of reaction of the two adjacent ions on such a model will be given by (19)

$$R = \frac{\omega}{\pi} n e^{-KA_c^2/2kT} \quad (6)$$

where ω is the vibrational frequency of that mode which allows the molecules to approach, K is the harmonic force constant of the mode (a harmonic potential is assumed). A_c is the amplitude of vibration at which the molecules are separated by a distance which allows them to interact. Note that because of compression of the lattice A_c will decrease with increasing shock pressure.

Because of the availability of data on the vibrational frequencies of ammonium nitrate and the relatively simple cubic structure of the high temperature phase, the model is applied to this material. The critical separation for reaction of two ammonium ions is taken as 2.92 Å which is twice the ionic radii of NH_4^+ . The reaction probability is assumed to be one when the ions achieve this separation. The reaction rate can then be calculated as a function of

shock pressure taking into account the decrease in A_c with shock compression of the lattice. Sufficiently high reaction rates as a function of shock pressure are obtained. For example, at 100 Kbs, the model predicts a rate of 10^5 s^{-1} . Such a mechanism could well be a possible way in which reactions are initiated in the solid state. It must be emphasized that the assumption of contact of ionic radii for critical separation is an arbitrary condition for reaction. The spirit of the calculation is to assess the feasibility of a collisional model of reaction in the solid state, particularly in a shock compressed solid. Another possibility that has been considered by other workers is that rotation of say CH_2 groups could enable hydrogen atoms and adjacent oxygen atoms of nitroamine explosives to interact (20).

MECHANICAL PROCESSES:

1. Molecular Deformation: As a shock pulse passes over a molecule of the lattice the molecule undergoes a time dependent compression. A mechanical breaking of the N-N bond of RDX could occur if the time dependent perturbation deforming the molecule causes a vibrational excitation to either points C in Figure 2, or directly causes an electronic excitation to B. Formal analysis of such mechanisms have been made. Assuming a Gaussian shaped shock pulse having rise time τ , the rate of excitation from the ground state to a vibrational state $N=L$ where bond breaking can occur is obtained to be

$$K = C_{OL} \frac{h}{2} m \omega^2 \Delta^2 \pi e^{-\omega^2 \tau^2/2} \quad (7)$$

where ω is vibrational frequency of bond, Δ the maximum deformation of bond, C_{OL} is the factor which determines probability for multiple excitations to state L, where the vibrational energy is equal to the dissociation energy. The critical parameters determining such a process are τ , the rise time of the shock, and Δ the amount of deformation of the bond. Equation (7) shows such a process is not possible unless τ is the order of magnitude of the period of vibration of molecule, (i.e. 10^{-13} s^{-1}). It is unlikely that this is possible. Recent molecular dynamic calculations of shock propagation in inert gas solids suggest larger rise times (21). Although rise times are not presently experimentally accessible, the amount of deformation Δ of the molecular bond for a given lattice compression can be estimated from experimental data. Using Raman spectroscopy and a high pressure diamond anvil pressure cell, the internal vibrational frequencies of the RDX molecule have been measured as a function of pressure up to 30 Kbs. The data shown in Figure 3 indicates very

small changes in the vibrational frequency with pressure. For example the frequency of the N-N vibration, the bond that is known to break by shock only increases by 0.3 percent at 30 Kbs. From a knowledge of the vibrational frequency of the bond and the dissociation energy a Morse potential can be constructed to relate bond energy to N-N separation. This can be used to estimate the rate of change of frequency with N-N separation. Coupled with the high pressure Raman data the change in N-N separation as a function of shock pressure can be obtained. The analysis shows that at 170 Kbs the equilibrium separation of the N-N bond would change insignificantly. Recent molecular orbital calculations of reaction surfaces of methylnitrate indicate that for unimolecular bond breaking to occur the molecule must be compressed by at least 5.0%. Such a change would result in much larger frequency shifts with pressure than have been measured in RDX. These results suggest that unimolecular bond breaking because of mechanical deformation of the molecule causing electronic or vibrational excitation is not possible.

2. Mechanical Vacancy Production: The abrupt mechanical deformation experienced by the lattice under shock compression will suddenly alter the potential that holds molecules to their lattice site. This time dependent change in the lattice potential could cause the molecules to be ejected from their lattice sites allowing molecules to approach each other, thereby enhancing reaction probability. The time dependent change in the potential may cause multiple vibrational excitations to an energy which allows the molecule to leave its site. The rate for this kind of a process can also be described by equation (7) where now Δ is the change in the lattice parameter of the solid which will, of course, be much larger than the change in the internal bond lengths of the molecule and the frequency will now be the vibrational frequency of the molecule in the lattice, which is an order of magnitude lower than internal molecular vibrations. The rate at which such a perturbation can remove a NH_4^+ ion from its lattice site in AN has been calculated as a function of shock pressure. Sufficiently large rates were obtained to account for a detonation process provided the rise time of the pulse is in the order of the vibrational period of the molecule at its lattice site. This mechanical model is really equivalent to the thermal process of ejection of a molecule from its lattice site. The time dependent change in the potential of the molecule at its lattice site causes excitations of lattice vibrations which is in effect shock heating. This mechanical model is really a fundamental explanation of how a shock pulse heats a lattice. The distinction then between a mechanical and thermal process for the vacancy production mechanism is artificial, and the general agreement between the calculated rates from the thermal approach and mechanical approach is to be expected.

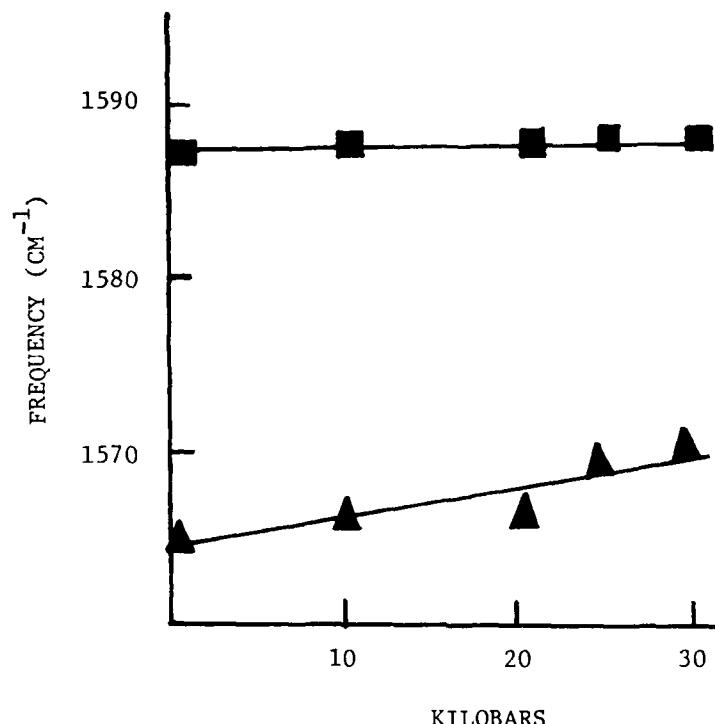


FIGURE 3. The internal vibrational frequencies of the N-N bond of the RDX molecule as a function of hydrostatic pressure applied to the RDX lattice.

CONCLUSION:

Analysis of macroscopic shock and thermal sensitivity data of solid explosives indicates that the chemistry caused by the shock pulse is a result primarily of shock heating but because of the rapid rise of temperature different reaction kinetics may occur than in slow heating.

A number of plausible molecular mechanisms by which a shock pulse may cause a chemical reaction in a solid were considered and expressions for reaction rates theoretically developed. The theories

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were used to calculate reaction rates as a function of shock pressure in real solid explosives. The results indicate that there is not sufficient time for energy to flow from the lattice to the internal states of the molecules of the lattice during shock compression, either due to thermal or mechanical means. In effect, the analysis shows that the internal states of the molecules are not in equilibrium with lattice states. This has two important implications. The reaction paths of slow thermal decompositon (where equilibrium is established) may not necessarily be the same as those occurring in shock heating. Also unimolecular chemical reactions are ruled out. In essence, an isolated RDX molecule should not detonate. The reactions of detonation are a result of cooperative processes between molecules of the lattice. This work indicates the most likely rate controlling process for initiation to detonation is the rate of collision between molecules of the lattice. Two possible ways in which collisions can occur in a shocked solid, large amplitude vibrations due to shock heating and removal of molecules from lattice sites were shown to be viable mechanisms.

Measurements of the effect of hydrostatic pressure on internal vibrational frequencies of the RDX molecule in the solid indicated that the deformation of the molecule is not large enough to be a mechanism of reaction. It was shown that for deformation to cause reaction, the deformation must occur in a time comparable to the vibrational period of a molecule, an unlikely situation. A summary of the probability of the various mechanisms of shock induced reaction considered is given in Table II.

The conclusion that the initial reactions leading to detonation are a result of cooperative interactions has important implications with respect to reducing sensitivity of existing formulations. To reduce sensitivity, the probability of collision of molecules in the solid must be reduced. This can be done by increasing the separation between the molecules in the solid by dispersing the molecules in chemically similar inert solids. The reaction rate should be proportional to the ratio of solvent to solute in the solid solution.

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Table II. Summary of Results of Analysis of Possible Mechanisms of Shock Induced Chemical Reactions in Solid Explosives

| Mechanism | Calculated Rate At Critical Pressure (s^{-1}) | Remarks |
|----------------------------|---|-----------------------------|
| THERMAL PROCESSES | | |
| Vibrational Relaxation | 10^4 | Unlikely |
| Electronic Excitation | 10^4 | Unlikely |
| Ionization | 0 | Unlikely |
| Vacancy Creation | 10^6 | A possibility |
| Large Amplitude Collisions | 10^5 | A possibility |
| MECHANICAL PROCESSES | | |
| Vibrational Relaxational | 0 | Not possible ($T > \tau$) |
| Vacancy Creation | 10^6 | possible |
| Electronic Excitation | 0 | Not possible ($T > \tau$) |

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